SQUID Picovoltometry of YBa₂Cu₃O₇ Single Crystals: Evidence for a Finite-Temperature Phase Transition in the High-Field Vortex State

P. L. Gammel, L. F. Schneemeyer, and D. J. Bishop *AT&T Bell Laboratories, Murray Hill, New Jersey 07974* (Received 13 August 1990)

Using a SQUID picovoltmeter, we have measured current-voltage curves of $YBa_2Cu_3O_7$ microtwinned crystals as a function of temperature in fields from 1 to 6 T. The data constitute evidence for a finite-temperature phase transition in the vortex state. Exponents derived within the framework of the vortex-glass model are found to be similar to thin-film values. Strongly temperature-dependent correlations in-volving up to 10^5 vortices near the transition, combined with the qualitative failure of thermal-activation models to fit our data, support the existence of a phase transition.

PACS numbers: 74.60.Ge, 74.70.Vy

The statics and dynamics of fluxoids in the mixed state of the high-temperature superconductors have generated intense interest and controversy. This is for two important reasons. First, an understanding of vortex interactions may help in obtaining high critical currents. Second, the mixed state is an excellent system in which to probe the decay of positional and orientational correlations and the nature of phase transitions in the presence of disorder.

The first evidence for unconventional behavior of the flux lattice in YBa₂Cu₃O₇ (YBCO) came from measurements of the decay of magnetization. An irreversibility line was found¹ which was associated with thermally activated depinning of single flux lines.² High-Q mechanical-oscillator measurements³ suggested a more controversial interpretation, namely, a melting transition from an ordered phase into a high-temperature flux liquid. The high temperatures, short coherence lengths, and large \hat{c} -axis anisotropies were postulated to increase the importance of thermal fluctuations to allow a melting transition resembling that seen in two-dimensional superconducting films.^{4,5} A phase transition in three dimensions is also predicted by Lindemann theories^{6,7} and models of entangled flux liquids.⁸

These simple phase-transition models fail⁹ to incorporate the disorder which leads to pinning. Larkin and Ovchinnikov¹⁰ showed that disorder leads to the destruction of long-range positional order in the flux lattice in all dimensions less than four. Hence, in three dimensions, pinning should dominate. An extensive literature explaining transport data in terms of thermally activated flux flow (TAFF) and flux creep has emerged.¹¹ These models postulate the existence of a finite resistivity for all T > 0 and only weakly temperature-dependent crossover currents for the onset of nonlinearities.

More recently, disorder has been included in theories which do involve a phase transition. Following the work of Ebner and Stroud,¹² Fisher, Fisher, and Huse¹³ have postulated a vortex-glass transition. Alternatively, Marchetti and Nelson¹⁴ have examined the entanglement model with disorder, drawing an analogy to polymer glasses. In both theories, a phase transition is signaled by the vanishing of the zero-frequency resistance. Koch *et al.*¹⁵ have analyzed thin-film I-V curves in terms of the vortex-glass model, although the strength of their scaling has been criticized.¹⁶ A phase with orientational order only—such as a hexatic—may also be stable in some part of the phase diagram.¹⁷ Worthington, Holtzberg, and Field¹⁸ have measured I-V curves on single-crystal YBCO which they use to argue for the presence of a hexatic at high temperatures with a quasi-first-order melting transition into an ordered phase at lower temperatures.

In this Letter we report on I-V curves of YBCO in fields of 1 to 6 T applied parallel to the \hat{c} axis with subpicovolt resolution. It was our reasoning that I-V curves at and near linear response at the picovolt level would provide the largest testable differences between the various theories. Our measurements of the temperature dependence of both the linear-response resistivity and the onset of nonlinear response strongly constrain theoretical fitting parameters and have allowed us to rule out a class of thermal-activation models.

In our experiment, voltages were measured using a modified BTI SQUID voltmeter.¹⁹ To adapt to the high temperatures involved, we used an input circuit consisting of NbTi wires soldered to a silver backbone and embedded in an epoxy and copper housing. The temperature gradient from 4 K to the sample temperature was designed to occur over the final 3 cm of the SQUID input circuit. The field gradients for the high-field magnet were engineered in conjunction with magnetic shielding to minimize the fields on the input circuit. With the sample chamber at 100 K, and an applied field of 6 T, the SQUID noise corresponded to a sensitivity of 1 pV/ $\sqrt{\text{Hz}}$. Owing to a contact resistance of $\sim 1 \Omega$ with this sample, the experimental noise was 7 pV/ \sqrt{Hz} . With a typical bandwidth of 0.01 Hz, this gave us a subpicovolt measurement capability.

The sample is identical to the one used in a previous

report by Palstra *et al.*¹¹ It is a thin slab 2 mm×1 mm×20 μ m with voltage leads 1 mm apart. The thin direction is the \hat{c} axis. The crystal is heavily twinned in the *a-b* plane. The zero-field superconducting transition is sharp in our measurement, with the resistance dropping by approximately two decades per 0.1 K over the range $0.5 > R/R_N > 10^{-6}$. The normal-state resistivity of 64 μ Ω cm is further evidence for the high quality of this sample. With a T_c in zero field of 88.033 K, however, the transition is suppressed from optimal. For $R/R_N > 10^{-2}$ the data are identical to those reported earlier.¹¹

The basic data consist of two types. At each field, a series of isothermal I-V curves, as shown in the inset to Fig. 3, constitute the bulk of the data. These curves were taken in two ways, which gave identical results. In the first, the voltage was measured at discrete current intervals. In the second, linear current sweeps of 1000 sec each were pieced together and averaged to cover the current range. In both types of measurement, the effective system bandwidth was 0.01 Hz. A second type of measurement, in which the current was square wave chopped at 0.001 Hz, with lock-in detection, was used to determine the linear-response value of the resistance for those data sets where the I-V curves showed a linear range. This value was always equal, within experimental errors, to the value obtained by fitting the linear part of the I-V curve. The current range over which this was applicable forms an integral part of the discussion of the data below.

We will focus on the data at 6.0 T. Data at other fields are similar, except as explicitly noted. The curves in the inset to Fig. 3, separated by approximately 0.1 K, show a crossover from linear to nonlinear behavior as the current is increased. In Fig. 1, the crosses indicate the



FIG. 1. The inverse logarithmic derivative of the resistivity should vanish in any model with a phase transition. The vortex glass is represented by the solid line. The dashed line is the prediction of the TAFF model as formulated by Griessen (Ref. 16). Inset: The power-law slope of the I-V curve for two voltage ranges, $10^{-8} < V < 10^{-7}$ (O) and $10^{-7} < V < 10^{-6}$ (\bullet).

temperature dependence of the linear-response data. The inverse logarithmic temperature derivative of R/R_N is displayed, where R_N is the linear extrapolation with temperature of the normal-state resistance. This quantity has a straightforward interpretation within several models of fluxoid dynamics and will be used as a test of their validity. A linear plot of R/R_N vs T shows the same qualitative features, including a shoulder at R/R_N ~ 0.2 , as noted previously.¹⁸ The dashed line is our simulation of the TAFF model suggested by Griessen.¹⁶ In his model, complexity is introduced in two ways. First, the sample is envisioned as a parallel summation of channels with different activation energies, with a distribution defined by a log-normal curve. In addition, each channel is shunted by a Bardeen-Stephen term. Despite this, the dashed line in Fig. 1, which arises from the linear-response part of our simulations²⁰ of his theory, is indistinguishable from e^{-U/k_BT} . There is clear disagreement with the data. The disagreement with other TAFF models¹⁶ is equally severe.

However, a good fit to our data is found within the framework of the vortex-glass model.¹³ In the scaling regime of this model, the resistance should vanish as $R \sim (T - T_g)^{\nu(z-1)}$. This implies that the plot shown in Fig. 1 of $(\partial \ln R/\partial T)^{-1}$ vs T should be a straight line which extrapolates to zero at T_g with a slope $1/\nu(z-1)$. The data shown in Fig. 1 give $T_g = 74.0$ K and $\nu(z-1) = 6.5$. Another way of fitting these data, shown in Fig. 2, is $\log_{10}(R/R_N)$ vs $\log_{10}(T - T_g)$. For a proper choice of T_g , the solid points should lie on a straight line with slope $\nu(z-1)$. The values thus determined are consistent with Fig. 1 and give overall $T_g = 74.0 \pm 0.2$ K and $\nu(z-1) = 6.5 \pm 1.5$. Deviations from scaling set in ~ 3 K above T_g . For other fields, not shown, the width of the



FIG. 2. Fits to the vortex-glass theory for the 6-T data. The solid symbols are the linear resistivity with $R_N = 0.064 \ \Omega$. The open symbols are the current where V/IR = 2 with $J_0 = 2.5 \times 10^3 \text{ A/m}^2$.

scaling region is reduced roughly in proportion to $T_c - T_g$, where T_c is the zero-field transition temperature.

For the nonlinear data, the simplest fit is again shown in the inset to Fig. 1. The average power-law fit to the *I-V* curve over the ranges $10^{-6} > V > 10^{-7}$ (solid symbols) and $10^{-7} > V > 10^{-8}$ (open symbols) is shown. As the voltage scale is reduced, the deviations from linearity are pushed to lower temperature and become more abrupt. For vortex-glass scaling, at $T = T_{g}$ one has $V \sim J^{(z+1)/2}$. While extrapolation with temperature to T_g is difficult, we estimate $z = 3.4 \pm 1.5$ from these data, comparable to what is presented below. We define a crossover current J_{sc} from V/IR = 2. Normalizing to $J_0 = J_{sc}(74.811 \text{ K}) = 2.5 \times 10^3 \text{ A/m}^2$, the data are shown in Fig. 2 as the open symbols. The onset of nonlinearities with current defined by a TAFF model is simply $J_{sc} \sim T$, which is a constant over the range of Fig. 2 and clearly disagrees with the data. Within the vortex-glass model the crossover current vanishes at the transition as $J_{\rm sc} \sim (T - T_g)^{2\nu}$, which is the straight line shown in the figure. This gives $v = 2.0 \pm 1$. Note the remarkable temperature dependence of J_{sc} , which varies by a factor of \sim 50 over less than 3 K in temperature at 75 K. This is extremely difficult to generate within a flux-creep or flux-flow scenario. Following Fisher, Fisher, and Huse,¹³ we may use this current to define a length scale via $\xi_d = (ck_B T/\phi_0 J)^{1/2}$. This is the length scale over which the current J can affect thermal distributions. At our lowest temperature $\xi_d \sim 15 \ \mu m$ implying cooperative motion of $\sim 10^5$ vortices. This length scale is almost a factor of 50 larger than that extracted by Koch et al.¹⁵



FIG. 3. The scaled data (points). For comparison the scaling function of Koch *et al.* (open symbols) and the prediction of TAFF (line) are shown. Inset: I-V curves at constant temperature for the 6.0-T data. The curves are spaced by approximately 0.1 K, excluding 74.377 K.

from their thin-film work. The exponent v from our nonlinear data can be combined with our linear-response data to give $v=2\pm 1$ and $z=4.3\pm 1.5$ within the framework of the vortex-glass theory. At a field of 4.0 T, Koch *et al.* found v=1.8 and z=4.7.

Within the vortex-glass theory, for $T > T_g$ the linearresponse resistance R and the crossover current J_{sc} can be used to scale all the I-V curves as shown in Fig. 3. For those data sets with V/IR > 4, R values could not be determined from the data, and were chosen to make the plot in Fig. 3 a smooth curve. These values were consistent with the vortex-glass scaling hypothesis for both the vanishing of the resistance and the current scale. Shown for comparison are the functional forms extracted from TAFF (solid line), which is simply $\sinh(J/J_0)$, and the scaling function found by Koch et al.¹⁵ (open circles). The curves were adjusted to agree at V/IR = 8. As already stated, the data show a significant qualitative departure from TAFF. Here, however, we also find differences from other data used to advocate a vortexglass transition. Three reasons could account for this departure. First, the thin-film data of Koch et al. focused on large nonlinearities. Their scaling was least accurate near linear response, where this comparison is made. Alternatively, both data sets may be correct, and the difference in ξ_d may represent a different scaling function for crystals as opposed to films. Of course, it may be that the dependence of the scaling function on the sample points to a failure of the vortex-glass hypothesis.

Shown in Fig. 4 is our composite phase diagram for the vortex state of YBCO. H_{c2} was taken from Welp *et al.*,²¹ shifted to agree with our zero-field T_c . The solid symbols are derived from fits with the vortex-glass hy-



FIG. 4. The phase diagram for the vortex lattice. H_{c2} is taken from estimates of Welp *et al.* T_c is taken from the zero-field resistive transition, which is sharp on this scale. The solid line is a $\frac{4}{3}$ power-law fit to the data. The dashed line is a linear fit.

pothesis. The prediction of the vortex-glass model for this phase diagram is shown by the solid line, $H_g \sim (T_c - T_g)^{4/3}$. This arises from the zero-field temperature dependence of the coherence length in the fluctuation regime. Mean-field theory would give a linear $T_g(H)$, which is shown by the dashed line. In this case, even near zero field there could be a transition below T_c . Such an effect has been noted in low-field magnetization data.²² More extensive low-field experiments are required to resolve this dilemma.

In conclusion, we have used a SQUID-based picovoltmeter to measure the I-V curves of microtwinned YBCO crystals at and near linear response. The data represent significant evidence for a finite-temperature phase transition in the vortex state. At our lowest temperature, the current scale for nonlinearities implies a coherence length $\xi_d \sim 15 \ \mu m$ for vortex motion. Such a coherence volume incorporates $\sim 10^5$ vortices, which strongly suggests that only many-particle theories with a phase transition will be able to explain our results. Our data significantly disagree with the predictions of present TAFF theories. Much of the data can be described within the framework of the vortex-glass model with $v=2\pm 1$ and $z=4.3\pm 1.5$. However, departures from earlier data with respect to the scaling function leave open questions as to the details of this model.

We would like to thank D. A. Huse, D. Nelson, D. S. Fisher, S. Coppersmith, A. F. Hebard, T. A. Worthington, B. Battlogg, and P. B. Littlewood for numerous helpful discussions. We would also like to thank R. Koch for sending us details of his fits to the scaling function and R. Griessen for responding to our questions about his thermal-activation model.

³P. L. Gammel et al., Phys. Rev. Lett. 61, 1666 (1988); R.

N. Kleiman et al., Phys. Rev. Lett. 62, 2331 (1989).

⁴B. A. Huberman and S. Doniach, Phys. Rev. Lett. **43**, 950 (1979); D. S. Fisher, Phys. Rev. B **22**, 1190 (1980).

⁵P. L. Gammel, A. F. Hebard, and D. J. Bishop, Phys. Rev. Lett. **60**, 144 (1988).

⁶M. A. Moore, Phys. Rev. B 39, 136 (1989).

⁷A. Houghton, H. A. Pelcovits, and A. Sudbo, Phys. Rev. B 40, 6763 (1989).

⁸D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); D. R. Nelson and H. S. Seung, Phys. Rev. B **39**, 9153 (1989); D. R. Nelson, J. Stat. Phys. **57**, 511 (1989); M. C. Marchetti and D. R. Nelson, Phys. Rev. B **41**, 1910 (1990); L. Xing and Z. Tesanovic, Phys. Rev. Lett. **65**, 794 (1990).

⁹E. H. Brandt et al., Phys. Rev. Lett. 62, 2330 (1989).

¹⁰A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).

¹¹T. T. M. Palstra *et al.*, Phys. Rev. Lett. **61**, 1662 (1988); M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988); M. Inui, P. B. Littlewood, and S. N. Coppersmith, Phys. Rev. Lett. **63**, 2421 (1989); C. W. Hagen and R. Griessen, Phys. Rev. Lett. **62**, 2857 (1989).

¹²C. Ebner and D. Stroud, Phys. Rev. B **31**, 165 (1985); S. John and T. Lubensky, Phys. Rev. B **34**, 4815 (1986).

¹³M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).

¹⁴M. C. Marchetti and D. Nelson, Phys. Rev. B **42**, 9938 (1990).

¹⁵R. H. Koch *et al.*, Phys. Rev. Lett. **63**, 1511 (1989); R. H. Koch, V. Foglietti, and M. P. A. Fisher, Phys. Rev. Lett. **64**, 2586 (1990).

¹⁶R. Griessen, Phys. Rev. Lett. **64**, 1674 (1990); S. N. Coppersmith, M. Inui, and P. B. Littlewood, Phys. Rev. Lett. **64**, 2585 (1990); S. Martin and A. Hebard (to be published).

¹⁷C. A. Murray et al., Phys. Rev. Lett. 64, 2312 (1990).

¹⁸T. K. Worthington, F. Holtzberg, and C. A. Field, Cryogenics **30**, 417 (1990).

¹⁹Biomagnetic Technologies Inc.

²⁰The dashed line in Fig. 1 is calculated using Griessen's expression (Ref. 16) with B = 6.0 T, $U_0^* = 85$ meV, $\gamma = 1.4$, $A = 3 \times 10^{-10}$, $S_0 = 4 \times 10^{-7}$, and $\rho_n = 6.4 \times 10^{-7}$ Ω m. For $U_0^* = 30$ meV, a more realistic value, the disagreement with the data is even more severe.

²¹U. Welp et al., Phys. Rev. Lett. 62, 1908 (1989).

²²H. Safar et al. (to be published).

¹K. A. Muller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987); A. P. Malozemoff *et al.*, Phys. Rev. B **38**, 7203 (1988).

²Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).